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Review article

Recent development of ionic liquid membranes

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Abstract

The interest in ionic liquids (IL) is motivated by its unique properties, such as negligible vapor pressure, thermal stability, wide electrochemical stability window, and tunability of properties. ILs have been highlighted as solvents for liquid–liquid extraction and liquid membrane separation. To further expand its application in separation field, the ionic liquid membranes (ILMs) and its separation technology have been proposed and developed rapidly. This paper is to give a comprehensive overview on the recent applications of ILMs for the separation of various compounds, including organic compounds, mixed gases, and metal ions. Firstly, ILMs was classified into supported ionic liquid membranes (SILMs) and quasi-solidified ionic liquid membranes (QSILMs) according to the immobilization method of ILs. Then, preparation methods of ILMs, membrane stability as well as applications of ILMs in the separation of various mixtures were reviewed. Followed this, transport mechanisms of gaseous mixtures and organic compounds were elucidated in order to better understand the separation process of ILMs. This tutorial review intends to not only offer an overview on the development of ILMs but also provide a guide for ILMs preparations and applications.

Keywords: Ionic liquid membrane; Supported ionic liquid membrane; Qusai-solidified ionic liquid membrane; Stability; Application

1. Introduction

Room temperature ionic liquids (RTILs) which are a type of molten electrolyte ($T_{melting}$ < 373 K), exhibit many attractive properties, such as negligible vapor pressure, non-flammability, high thermal stability, a liquid range of up to at least 300 °C, solubility of a wide range of organic and inorganic compounds, and wide electrochemical window in contrast to the conventional molecular solvents [1–3]. Moreover, their physicochemical properties can be tailored to satisfy specific chemical tasks by the appropriate selection of anion, cation, and substituents on the cationic constituent. The structures of some commonly used cations and anions of ILs are shown in Fig. 1, where the R_1, R_2, R_3 and R_4 groups are alkyl groups. Therefore, some encouraging results have been achieved with respect to their utilization as a benign medium or solvent in a wide variety of applications, *e.g.* separation of various compounds [4–8], extractions [9–13] and chemical reactions [14–18].

However, some limitations of IL, *e.g.* the high price for synthesis and high energy requirement for recycling, could affect the economic viabilities in some potential processes. These drawbacks might be overcome by making use of ionic liquid membrane (ILM) technology, which consists of the feed and permeate phases separated by membrane containing IL allowing simultaneous extraction and stripping at each side of ILM. For an ILM, IL can be stabilized by either impregnating it inside the pores of the support membrane or quasi-solidification it to endow material with good mechanical strength. ILM techniques require fairly less amount of IL as carrier, and would not require additional steps for the

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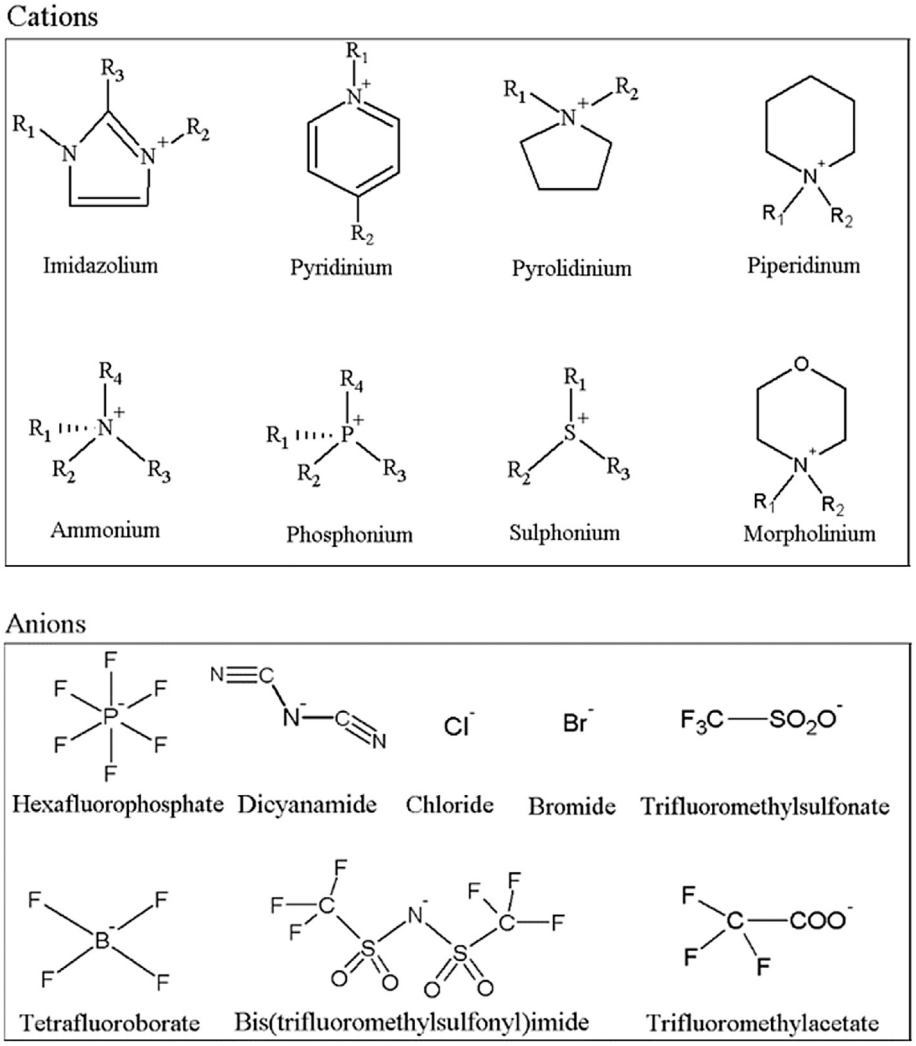

Fig. 1. Commonly used cations and anions of ionic liquids.

recycling of IL. Thus, ILMs have many advantages, such as easy fabrication of flexible and compact devices, low energy requirements, low capital and operating costs, *etc*. In addition, due to the unique properties of IL, *e.g*. negligible vapor pressure and high viscosity, ILMs are more stable in comparison with the traditional supported liquid membranes (SLMs) based on organic solvents [19–32]. Therefore, ILMs show promising application potential.

During the last decade, some investigators have reported that ILMs were used to implement the separation of different compounds (*e.g*. alcohols [33–37], gases [38–51], organic acid [52–56], esters [57,58] and aromatic hydrocarbons [59,60]), to fabricate advanced electrochemical devices (*e.g*. lithium batteries [61,62], fuel cells [63,64] and solar cells [65–67]), to facilitate catalytic reactions [68], *etc*. An overview of some fields for utilizing ILMs is given in Fig. 2. Among these applications, a large number of publications relating to the separation of various compounds indicate that research in this field was growing dramatically. The research progress of gas separation, especially CO_2 separation, has been addressed in depth. For example, Dai et al. [69] presented the recent progress on CO_2 separation membranes based on ILs in detail. In addition, Lozano et al. [70] have also given the recent advances in supported ionic liquid membranes (SILMs).

For successful separation of mixtures through ILMs, there are two key requirements: a good selection of the carrier and the establishment of stable ILMs which can withstand the long term operation. This review was mainly dedicated to the recent development in ILM technology, including preparation methods, stability, application of ILMs, as well as transport mechanisms. The preparation methods of ILMs were elaborated in detail. The influence of components properties, preparation method of ILMs, and compatibility between IL and support membrane/gel, *etc*. on the stability of ILMs was highlighted. Recent developments of ILMs in the separation of different chemical species, such as mixed gases, organic compounds and metal ions, were also discussed.

2. Preparation methods of ILMs

According to the immobilization approach of IL, ILM can be mainly divided into the following kinds, *i.e*. supported ionic

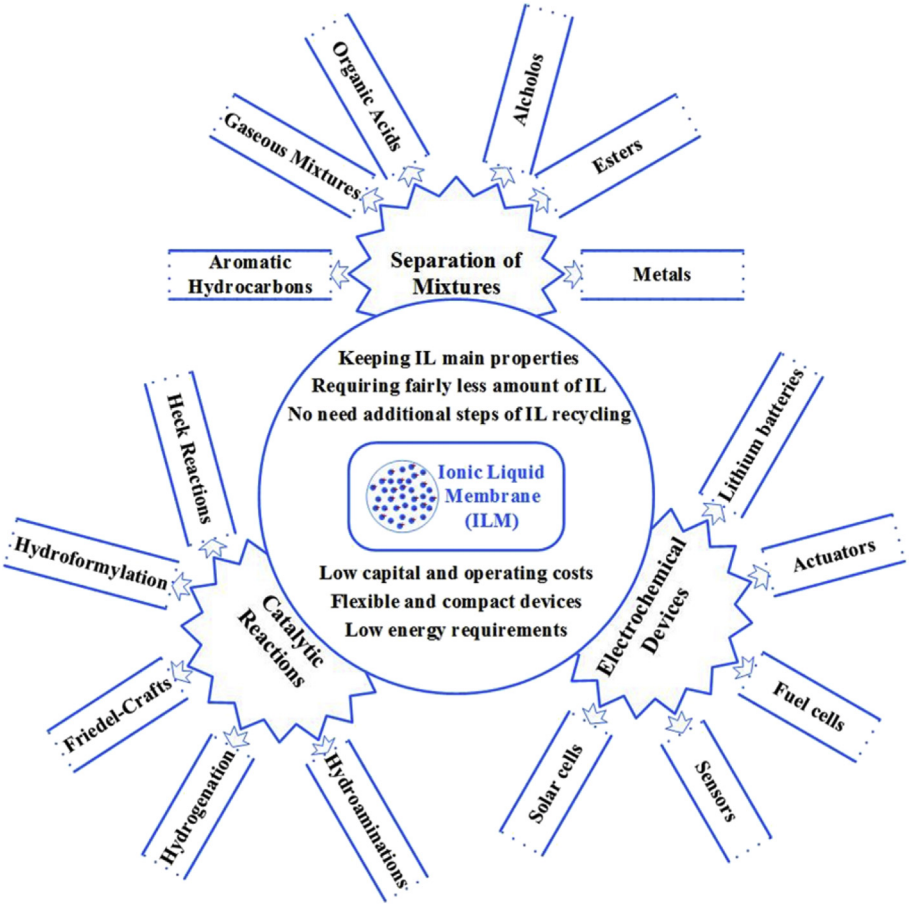

Fig. 2. Properties and applications of ionic liquid membranes.

liquid membrane (SILM) and quasi-solidified ionic liquid membrane (QSILM) which were described as follows in detail. Two main approaches for utilizing ILs as membrane materials are shown in Fig. 3.

2.1. Supported ionic liquid membrane

A SILM is one of the three phase liquid membrane system in which the IL is held by capillary forces in the pores of support material. The support materials mainly include polymeric and inorganic membranes. In general, there are three methods for the preparation of SILM, *i.e.* direct immersion, vacuum and pressure, which can play an important role on the operation performance of SILM due to the relatively high viscosity of IL.

For the direct immersion method, immobilization of IL takes place by soaping the support material in the IL under the ambient pressure. Then, the excess IL should be removed from the material surface either by leaving to drip overnight or by wiping up softly with a tissue. This method is the easiest way

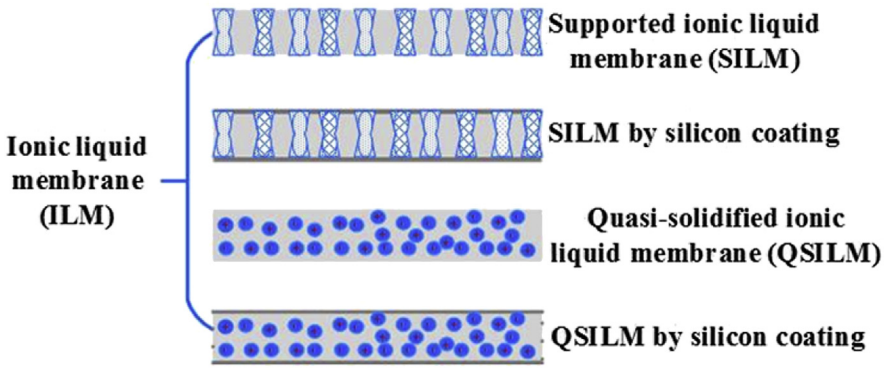

Fig. 3. Two main approaches for utilizing ILs as membrane materials.

in comparison with the other two methods. Some SILMs have been prepared using this method [71–75]. For example, by soaking a hydrophilic PVDF membrane in six phosphonium-based ILs, several SILMs were obtained [71]. Miyako et al. [72] reported that a SILM was obtained by immersing a hydrophobic PP film into [BMIM]PF_6. Hashim and coworkers [73] mentioned that SILMs were prepared by impregnating a 100% solution of ILs, *i.e.* eight imidazolium-, one phosphonium- and one ammonium-based ILs inside the different membranes (*i.e.* Fluoropore, Durapore, Mitex). For the separation of organic compounds, the prepared SILMs using this method are generally suitable for the transport experiments [71–75].

For the vacuum method, the SILMs can be achieved as follows. Firstly, the support material is placed inside a vacuum-tight chamber for a certain time in order to remove air from the pores of the material. Then, IL is spread out at the membrane surface while keeping vacuum in the chamber. Finally, the excess IL on the surface of membrane should be removed. Some polymeric and ceramic nanofiltration (NF) membranes have been used as support materials to immobilize IL using this method [76–82]. For example, Fortunato et al. [79] prepared SILMs using four hydrophilic membranes with equal nominal pore size, *i.e.* PP, PVDF, nylon and PES as support membranes and two ILs, *i.e.* [BMIM][PF_6] and [OMIM][PF_6] as liquid phases. They found that the amount of immobilized IL decreased with an increase in viscosity of the IL due to the decrease of the capillary force. For the separation of organic mixtures, the SILMs prepared using this method can be utilized for the pervaporation experiments under lower pressure.

For the pressure method, the immobilization of IL is carried out by the following steps: (1) placing the material in an ultrafiltration unit, (2) adding an amount of IL in the unit, (3) applying a certain nitrogen pressure to force the IL to flow into the pores of the material, (4) releasing the pressure once a thin layer of IL was apparent on the surface of the membrane, and (5) removing the excess IL on the membrane surface using the same way as the last step of direct immersion method. The amount of IL immobilized by this method is likely to be independent of the IL. For example, SILMs with [BMIM][Cl], [BMIM][BF_4] or [BMIM][NTf_2] supported in Nylon organic membrane were prepared using pressure method [83], and the resulting SILMs confirmed that the pores of support material were completely filled with each of the ILs. The prepared SILMs using this method can be used for the pervaporation experiments even under higher pressures [82–85].

2.2. Quasi-solidified ionic liquid membrane

QSILM can be obtained by casting a solution containing an IL and a special gel to form a thin and stable quasi-solidification film. Compared to the preparation methods of SILM, quasi-solidification of IL is a promising method to effectively prevent its leakage by the self-assembly of organic molecules. QSILMs have been employed as highly selective separation membranes. The current gels used to quasi-solidification of IL were mainly polymers and low-molecular-weight organic gelators (LMOGs).

For polymers, PDMS is a candidate for use as the material to quasi-solidify IL due to its high thermal and chemical stability. However, the prepared QSILMs have been of limited utility because PDMS is immiscible with many ILs. Izák et al. [86] prepared the quasi-solidified [BBIM]BF_4-PDMS membranes containing 0, 10, 20 or 30 wt.% of [BBIM]BF_4. The QSILMs contained amorphous and crystalline phases of PDMS and a dispersed phase of [BBIM]BF_4 because the two components were not compatible. In addition, by covalent binding a Tf_2N-based IL into the PDMS backbone polymer, an IL-poor QSILM was obtained because of the limited miscibility of PDMS in IL [87]. To overcome the immiscibility of PDMS in IL and increase IL loadings, Horowitz et al. [88] successfully prepared a QSILM with IL loadings of up to 80 wt.% by casting a resin, which was obtained by stirring a mixture of a functionalized PDMS oligomer, formic acid and an IL. The used ILs included [EMIM][TFSI], [BMP][TFSI] and [EMIM][TCB]. Besides PDMS used to quasi-solidify IL, some other polymeric materials, *e.g.* PVDF-HFP, PVC, cellulose polymer, PEG, VP, VPrr and PI have also been utilized for the preparation of QSILMs [89–102]. Carlin et al. [89,90] synthesized firstly a gas-permeable IL-polymer gel with PVDF-HFP copolymer of more than 30 wt.%, imidazolium-based IL and 2-methylpentene as helper additives. Matsumoto et al. [96] prepared the QSILMs by dissolving ILs, *i.e.* Aliquat 336, Cyphos 101, Cyphos 102 and Cyphos 104 in PVC. Only an anisotropic viscous solution rather than a gel was obtained by dissolving a considerable amount of cellulose polymer (more than 10 wt.%) in the [BMIM][Cl] [97]. However, in the same research group, a quasi-solidified [HMIM][NTf_2]-PEG membrane was successfully obtained by gelation of the [HMIM][NTf_2] using the cross-linking of tetra-armed PEG amine and bifunctional PEG succinimide oligomers [98]. This result indicated that the noncoordinating, hydrophobic anion-based ILs could be quasi-solidified by PEG under certain experimental conditions. Wand et al. [100] found that the binary system of PI and IL behaved as a compatible system when the IL content was below 35 wt.%. Therefore, a QSILM composed of PI and [BMIM][NTf_2] IL can be successfully prepared by a casting method.

For LMOGs, there are a limited number of gelators that can quasi-solidify ILs [103–111]. An QSILM can be obtained as follows: 1) mixing a small amount of LMOG (2–5 wt.%) with an IL at elevated temperatures; and 2) casting the mixture on a glass substrate. The commonly used LMOGs include 12-hydroxystearic acid, cyclo (L-β-3,7-dimethyloctylasparaginyl-L-phenylalanyl) and cyclo (L-β-2-ethylhexylasparaginyl-L-phenylalanyl), especially 12-hydroxystearic acid. Voss and coworkers prepared a QSILM based on physical gelation of [HMIM][NTf_2] with 12-hydroxystearic acid [103]. The maximum loading of LMOG in IL was found to be approximately 1.5 wt.%, and the resulting QSILM has good mechanical strength while maintaining liquid-like gas transport properties similar to that of the neat [HMIM][NTf_2]. Plaza et al. [104] obtained a

solution by dissolving 12-hydroxystearic acid in the [BMIM]PF_6 at 80 °C. When the solution was cooled down to the room temperature, a gel was obtained through a network formed by either hydrogen-bonding and/or van der Waals forces in the LMOG and IL. To avoid excessive cross-linking reactions and ensure the mechanical stability of the IL gel, the concentration of 12-hydroxystearic acid should be kept at a relatively low value. Hanabusa et al. [105] reported special LMOGs that could quasi-solidify a wide variety of ILs even without helper additives. Among these LMOGs, cyclo (L-β-3,7-dimethyloctylasparaginyl-L-phenylalanyl) and cyclo (L-β-2-ethylhexylasparaginyl-L-phenylalanyl) were excellent gelators for a wide range of RTILs. The solubilities of the two LMOGs in ILs are high enough so that they do not require a cosolvent to facilitate dissolution. Furthermore, they have the potential to provide solid-like materials with an ionic conductivity approaching that of a pure IL. Besides, several other gelators, which showed good quasi-solidification capability in a limited range of ILs, were also used to prepare QSILMs. Using a LMOG consisting of glycolipid and L-glutamic acid, Kimizuka et al. [106] studied physical gelation of ILs, but the gelation ability of the LMOG seemed to be limited to the imidazolium-based ILs with Br ion. Amaike et al. [107] synthesized a LMOG *via* three reaction steps from cholesteryl chloroformate and 4-nitrophyeyl-β-D-glucopyranoside. Due to the fact that this kind of LMOG is hardly soluble in ILs, the acetone has to be added to solubilize the gelator. A cholesterol-based LMOG has also been applied to physical gelation of ILs [108]. By addition of a small amount of gelator, imidazolium- and pyridinium-based ILs are easily quasi-solidified to be the sufficiently thermo-stable gels. In addition, a QSILM, which can be fabricated to be dye-sensitized solar cells, was synthesized by dissolving N-benzyloxycarbonyl-L-isoleucylaminooctadecane in imidazolium-based ILs with iodide anion [109]. Although this gelator is an all-powerful LMOG for ordinary solvents [110], its quasi-solidification ability toward ILs is only confined to the imidazolium-based ILs with bromide or iodide.

3. Stability of ILMs

Compared to the organic solvents SLMs, ILs possess many attractive characteristics, such as negligible vapor pressure, higher viscosity and tunability of properties through the change in the combination of cation and anion. Therefore, ILM has shown great potential as an alternative to the conventional SLM in separation processes because ILM technique offers the advantages of high selectivity (by using a task-specific IL) and negligible loss of IL as liquid phase under a cross-membrane pressure gradient [112].

3.1. Stability of SILM

In the case of SILM, its stability is mainly affected by the properties of the support membrane and IL, preparation method of SILM, compatibility between IL and support membrane, as well as interfacial tension between the aqueous and membrane, *etc*. [113–115].

The properties of the support membrane, such as type, pore size or porosity play an important role in the stability of SILMs. For the type of support membrane, a slight increase of [BMIM][PF_6] concentration in aqueous phase was observed for the SILM with nylon membrane [116]. Whereas, the concentration of [BMIM][PF_6] remained essentially constant for the SILMs with PP, PVDF or PES. In addition, based on a hydrophobic IL, SILMs with a hydrophobic membrane were more stable than those prepared with a hydrophilic membrane due to a possible weak interaction between the hydrophilic support membrane and the hydrophobic IL [116,117]. Moreover, the IL loss of SILMs could be mainly attributed to membrane compression and IL extrusion from the large pores [118]. Therefore, the SILMs system can tolerate a relatively high transmembrane pressure by choosing a proper pore size of support membrane. In general, membranes with a pore size of 100–200 nm are suitable for the preparation of SILMs [119]. To further enhance the SILM pressure resistance, avoid the pitfall of IL instability associated with microporous membranes and reduce the possibility of swelling of the porous resulted from IL in SILMs, some inorganic NF membranes have also been used to prepare SILMs. Compared to the polymeric membranes, the inorganic NF materials as support membranes has shown a great potential due to their excellent thermal stability, mechanical strength and long lifetime, although only a few reports can be found [78,85,120–134]. The SILMs based on a special inorganic NF membrane were stable under a transmembrane pressure up to 10 bar [85]. Kulkarni et al. [78] found that ILs with [DCA] anion did not discharge from the porous ceramic NF membrane structure made of TiO_2 or Al_2O_3 even at temperatures up to 200 °C. It can be concluded that SILMs based on ceramic NF supports might be used for practical applications at relatively high temperatures and pressures. For porosity of support membrane, high porosity is generally favorable because it can support the IL under hydrostatic pressure which increases the stability. Also, the transport efficiency can be enhanced by providing more surface area.

The properties of IL can be tailored to specific application by changing either the cation or the anion in IL. Therefore, the proper selection of ILs can greatly increase the stability of SILMs. Besides hydrophobicity and surface tension, viscosity of IL plays an important role in the stability of the SILM. ILs with high viscosity are not easily removed from the pores under a cross-membrane pressure difference as they are immobilized by large Van der Waals forces [123,124]. Whereas ILs with low viscosity were prone to be displaced from the pores of the support.

Preparation method of SILM is also one of the critical issues for membrane stability. Hernández-Fernández and coworker [83] found that the losses of ILs generally increased with an increase in viscosity of IL immobilized inside the pores of support membrane by vacuum method. The main problem lies in the fact that less ILs are immobilized in the internal layers of the support materials due to the higher

capillary force for more viscous ILs. This can be mainly attributed to the fact that ILs with high viscosity are mainly immobilized on the most external layer of the support membrane by vacuum method, and thus are easily displaced into the aqueous phase during operation. However, the SILMs with the highest stability can be obtained by pressure method, and small losses of IL was observed even after 7 days of operation. Furthermore, the amount of IL was possibly independent of the IL when immobilization was carried out under pressure [83]. Therefore, to ensure that all the pores of support are filled with ILs to obtain the stable SILMs, the methods of direct immersion and vacuum are suitable for the low-viscosity ILs, whereas for the high-viscosity ILs, pressure method is preferred.

In addition, the compatibility between IL and support membrane has an influence on the states of the ILs in the support pores. The electrostatic interaction between the organic cations in ILs and the charged functional group on the surface of the support membrane affects the stable performance of SILMs [79]. This is mainly due to the fact that the organic cations in ILs have unique molecular structure, polarity and charge distribution throughout the imidazolium ring. In addition, due to the exist of the interfacial tension between aqueous and membrane phase, the water microenvironments inside the IL phase of SILM, which constitute a non-selective environment for solute transport, may be formed during the transport of aqueous solution because of the water solubility of IL [82,125]. This will result in a deterioration of the SILM performance, and even a loss of IL from SILM due to the decrease of its viscosity.

3.2. Stability of QSILM

QSILMs are attractive because they are easier to prepare membrane modules for industrial applications, and can effectively control the flowing of ILs away from the resulting membranes. Therefore, the operational stability of QSILM is higher than that of conventional SILM from which IL might be displaced at relatively low cross-membrane pressure or high temperature during the process of pervaporation. The mechanical strength and thermal stability of QSILM are mainly affected by the concentration of gel or IL, as well as the compatibility of gel with IL.

The concentration of gel or IL, is an important parameter contributing to the stability of QSILM [105,126–130]. Chen and coworkers [126] studied the effect of IL content on the mechanical properties of QSILM made from PVDF and [EMIM][$B(CN)_4$]. Although the Young's modulus, tensile strength and extension at break decreased with an increase in IL loading, these QSILMs were still very stable at a differential pressure range of up to 5 atm. Jansen et al. [127] found that the melting point, elastic modulus and break strength of QSILM decreased with increasing imidazolium-based IL content. The elastic modulus of QSILM containing only 20 wt.% of the IL decreased one order of magnitude compared with the neat p (PVDF-HFP) copolymer. However, a high-performance QSILM can be prepared using the tetra-armed PEG ion gel containing a large fraction of IL (94 wt.%), showing excellent thermal stability over temperatures up to 100 °C [128]. In addition, the strength of QSILM based on cyclo (L-β-3,7-dimethyloctylasparaginyl-L-phenylalanyl) or cyclo (L-β-2-ethylhexylasparaginyl-L-phenylalanyl) LMOG increased at a rate nearly proportional to the concentration of added LMOG [105]. The resulting QSILMs were very thermally stable mainly due to the fact that the three-dimensional networks consisting of highly intertwined fiber were built up by the noncovalent interactions. Nguyen and coworkers [129] also reported that the sol-gel transition temperature and mechanical stability of QSILM based on cyclo (L-β-3,7-dimethyloctylasparaginyl-L-phenylalanyl) could be improved by increasing the LMOG loading. It can be concluded from above that, in the most cases, the maximum breaking strength and the melting point of the QSILM will increase with a decrease in IL loading.

The compatibility of gel with IL which is highly important to the stability of QSILMs, can be affected by the characteristics of components [129–133]. The thermal stability of the QSILM decreased with an increase in the length of alkyl chain attached to the imidazolium cation probably due to the fact that IL with the long alkyl chain shows less compatibility with the polymer matrix and tends to give significant phase separation [130]. It was indicated that p(PVDF-HFP) copolymer are compatible with the imidazolium-based ILs with [NTf_2] anion, especially ILs with short alkyl chain [129]. However, the burst pressure for the QSILM made from [HMIM][NTf_2] is higher than that from [EMIM][NTf_2] because the longer alkyl chain attached to the imidazolium cation causes stronger interactions with the aspartame-based LMOG molecules. The QSILM prepared by simple dissolution of [C_{10}MIM][TCB] in PEBA is unstable even if it is possible to dissolve up to 55 wt.% of IL in PEBA [36]. During pervaporation, about half of the IL immobilized in the PEBA was leached out of the QSILM. The QSILMs consisting of PVC main chains and polymerized ionic liquid (PIL) side chains exhibit good mechanical properties without losing separation properties due to the microphase-separated structures with molecular self-assembly [131]. The QSILMs made from two thermo-stable polymeric materials, *i.e.* PMDA-ODA PI and PBI, can maintain excellent mechanical and thermal stability over a range of transmembrane pressure (2–6 bar) and temperatures (100–200 °C) [132]. The quasi-solidified [P_{4444}][Pro]-PDMAA membrane ruptured at an applied pressure of less than several hundred kPa, whereas, quasi-solidified [P_{4444}][Pro]-PVP membrane sustained a stress of more than 1 MPa [133]. This results indicated that PVP has better compatibility with [P_{4444}][Pro] in comparison with PDMAA polymer.

4. Application of ILMs

The separation performance of ILM can be significantly enhanced using a special IL that was immobilized in the pores of a given support membrane or quasi-solidified into a thermally stable material by LMOG or polymer. In the last decade, ILMs have attracted great attention in the separation of mixtures, *e.g.* gases, metals, and organic compounds.

Table 1
Use of ILMs in separation of gases.

Application	IL	Support membrane/LMOG/ Polymeric materials	Reference
Separation of CO_2 from N_2	[APMIM][NTf_2]	Tubular Al_2O_3	[165]
	[EMIM][Ac]	Porous Al_2O_3/TiO_2 tubes	[164]
	[EMIM][Ac]	PVDF	[172]
	[EMIM][$B(CN)_4$]	PTEE	[155]
	[EMIM][BF_4]	PVDF	[168]
	[EMIM][$C(CN)_3$]	PTEE	[155]
	[EMIM][DCA]	PTEE	[155]
	[EMIM][ESO_4]	PTEE	[173]
	[EMIM][Gly]	PTEE	[169]
	[EMIM][MSO_4]	PTEE	[155]
	[EMIM][NTf_2]	PES	[160]
	[EMIM][NTf_2]	Al_2O_3-Anodisc	[119]
	[EMIM][NTf_2]	PTEE	[169]
	[EMIM][SCN]	PTEE	[155]
	[EMIM][TCM]	Silica Nanoporous Ceramic	[156]
	[EMIM][TFA]	Al_2O_3-Anodisc	[119]
	[C_3MIM][NTf_2]	PTEE	[169]
	[BMIM][Ac]	PVDF	[172]
	[BMIM][Ac]	Al_2O_3-Anodisc	[119]
	[BMIM][BETI]	PES	[160]
	[BMIM][BF_4]	PVDF	[117,140]
	[BMIM][BF_4]	PES	[157]
	[BMIM][DCA]	PVDF	[140]
	[BMIM][NTf_2]	PTEE	[167]
	[BMIM][NTf_2]	PVDF	[117,140,166,167]
	[BMIM][NTf_2]	Al_2O_3-Anodisc	[119,121]
	[BMIM][OTf]	PVDF	[140]
	[BMIM][PF_6]	PTEE	[167]
	[BMIM][PF_6]	PVDF	[117,140,166,167]
	[BMIM][TCM]	Silica nanoporous ceramic membrane	[156]
	[HMIM][NTf_2]	Al_2O_3-Anodisc	[119]
	[OMIM][PF_6]	PVDF	[117]
	[C_8F_{13}MIM][NTf_2]	Al_2O_3-Anodisc	[121]
	[C_{10}MIM][BF_4]	PVDF	[117]
	[P_{4444}][Gly]	PTEE	[169]
	[Vbtma][Ac]	PVDF	[172]
Separation of CO_2 from CH_4	[EMIM][$B(CN)_4$]	PTEE	[155]
	[EMIM][BF_4]	PES	[160]
	[EMIM][$C(CN)_3$]	PTEE	[155]
	[EMIM][CF_3SO_3]	PES	[160]
	[EMIM][CF_3SO_3]	PVDF	[160]
	[EMIM][DCA]	PES	[160]
	[EMIM][DCA]	PTEE	[155]
	[EMIM][ESO_4]	PTEE	[155]
	[EMIM][MSO_4]	PTEE	[155]
	[EMIM][NTf_2]	PES	[148,160]
	[EMIM][NTf_2]	PVDF	[148]
	[EMIM][NTf_2]	PA	[148]
	[EMIM][NTf_2]	PP	[148]
	[EMIM][NTf_2]	PVDF	[121]
	[EMIM][NTf_2]	PA	[148]
	[EMIM][NTf_2]	PP	[148]
	[EMIM][NTf_2]	PVDF	[148]
	[EMIM][SCN]	PTEE	[155]
	[C_3NH_2MIM][NTf_2]	PTEE	[135]
	[C_3NH_2MIM][CF_3SO_3]	PTEE	[135]
	[BMIM][BF_4]	PVDF	[135]
	[BMIM][NTf_2]	PTEE	[135]
	[BMIM][NTf_2]	PVDF	[117]
	[BMIM][NTf_2]	PES	[148]
	[BMIM][NTf_2]	PVDF	[148]

(*continued on next page*)

Table 1 (*continued*)

Application	IL	Support membrane/LMOG/ Polymeric materials	Reference
	[BMIM][NTf_2]	PA	[148]
	[BMIM][NTf_2]	PP	[148]
	[BMIM][PF_6]	Al_2O_3-Anodisc	[164]
	[BMIM][PF_6]	PVDF	[117]
	[BMIM][TfO]	PES	[148]
	[BMIM][TfO]	PVDF	[148]
	[BMIM][TfO]	PA	[148]
	[BMIM][TfO]	PP	[148]
	[HMIM][Tf_2N]	PES	[166]
	[OMIM][PF_6]	PVDF	[117]
	[C_{10}MIM][BF_4]	PVDF	[117]
	h[MIM]$_2$[Tf_2N]$_2$	alumina NF membrane	[173]
	pr[MIM]$_2$[Tf_2N]$_2$	alumina NF membrane	[173]
Separation of CO_2 from CH_4	styrene-based IL	PES	[158]
	styrene-based IL	PES	[158]
	styrene-based IL	PES	[158]
Separation of biohydrogen from CO_2 and N_2	[C_3NH_2MIM][CF_3SO_3]	PVDF	[140]
Separation of CO_2 from He	[HMIM][NTf_2]	PS	[163]
Separation of CO_2 from CO	[SMMIM][PF_6]	γ-alumina NF membrane	[50]
Separation of CO_2 from CH_4, N_2 and H_2	[EMIM][TFSI]	p (VDF-HFP)	[127,150]
Separation of CO_2 from CH_4 and N_2	[EMIM][Ac]	PVDF	[152]
	[EMIM][DCA]	PVDF	[152]
	[EMIM][Lac]	PVDF	[152]
	[EMIM][NTf_2]	PVDF	[152]
	[EMIM][SCN]	PVDF	[152]
Separation of CO_2 from gaseous mixtures of CH_4, O_2 and N_2	[HMIM][NTf_2]	PES	[137]
	[OMIM][NTf_2]	PES	[137]
Separation of SO_2 from air	[MIM][AC]	PVDF	[174]
	[BIM][AC]	PVDF	[174]
Separation of SO_2 from N_2	[N_{2224}][acetate]	PES	[177]
	[N_{2224}][diglutarate]	PES	[177]
	[N_{2224}]$_2$[dimaleate]	PES	[177]
	[N_{2224}][dimalonate]	PES	[177]
	[N_{2224}]$_2$[maleate]	PES	[177]
	[N_{2224}]$_2$[malonate]	PES	[177]
	[N_{2224}][NTf_2]	PES	[177]
	[N_{2224}][propionate]	PES	[177]
Separation of SO_2 from CH_4	[EMIM][BF_4]	PES	[175]
	[BMIM][BF_4]	PES	[175]
	[BMIM][PF_6]	PES	[175]
	[BMIM][Tf_2N]	PES	[175]
	[N_{2224}][acetate]	PES	[177]
	[N_{2224}][dimalonate]	PES	[177]
	[N_{2224}]$_2$[dimaleate]	PES	[177]
	[N_{2224}]$_2$[maleate]	PES	[177]
	[N_{2224}]$_2$[malonate]	PES	[177]
	[N_{2224}][NTf_2]	PES	[177]
	[N_{2224}][propionate]	PES	[177]
Separation of H_2S from CH_4	[BMIM][BF_4]	PVDF	[176]

4.1. Separation of gases

ILMs exhibited great advantages for the separation of gaseous mixtures in comparison to the conventional SLMs because IL showed the high solubility capacity of different gaseous species and was hardly lost through volatilization [134–143]. Table 1 shows commonly used support membrane/gel and their corresponding IL as well as application of ILM in

the separation of gases. In these works, the influence of some main factors, *i.e.* type and content of components used to synthesize ILM, water content and plasticization behavior of IL, temperature, as well as pressure on the separation performance of ILM were investigated.

4.1.1. Separation of CO_2

The separation of CO_2 from industrial gas mixtures has attracted worldwide interest due to global warming issue. Since the interaction between the quadrupole moment of CO_2 and the electrical charge of IL provides an enhancement in the solubility over other gases [144,145], ILMs have been proved to be very promising for the selective separation of CO_2 from gas mixtures, such as N_2, CH_4, H_2 and He. Some researchers have extensively investigated the effect of type and concentration of components on the CO_2 separation performance of ILMs [117,119,121,127,134,135,137,146–168]. Scovazzo et al. [146,147] studied the effect of anions in ILs on the performance of SILMs with a porous hydrophilic PES support for separation of CO_2/N_2 and CO_2/CH_4. It was found that the performance of SILMs with [DCA] anion-based IL is competitive or superior to other SILMs on the basis of Robeson' upper bound correlation for CO_2/N_2 and CO_2/CH_4. Neves et al. [117] investigated the effect of alkyl chain lengths of the IL cation and type of anions on the selectivities of SILMs for CO_2/N_2 and CO_2/CH_4 gaseous mixtures. The results indicated that the gas permeability for all gases increased with an increase in the alkyl chain length of IL cation and decreased with an increase in IL viscosity mainly affected by anion of IL. In addition, they also found that the nature of the support membrane strongly affected the transport rates and selectivity in the separation of mixed gases. SILM with [BMIM [NTf_2] immobilized in the hydrophobic PVDF support exhibited best performance for both separations of CO_2/N_2 and CO_2/CH_4, mainly due to the best affinity between the IL and the support. The impact of cation structure on mechanical properties of SILMs prepared by immobilizing [BMIM]-based ILs inside pores of polypropylene support has also been investigated by Cichowska-Kopczzynska et al. [148]. It was found that the mechanical stability of SILM decreased with an increase in the hydrocarbon chain of IL cation resulting in the more disruptions in SILM structure. The effect of confinement on gas sorption of CO_2 and H_2 was studied by confining [HMIM][NTf_2] into carbon nanotubes (CNTs) [149]. The IL molecules in the CNT exhibited self-diffusivity coefficients about 1–2 orders of magnitude larger than the corresponding bulk IL molecules. Sorption of CO_2 and H_2 gases in the composite material consisting of CNT and IL indicates that H_2 molecules diffuse about 1.5 times faster than the CO_2. In addition, to investigate the effect of IL content on the permeability of different gas species (*i.e.* CO_2, N_2, CH_4, H_2 and O_2), the QSILMs containing from 20 wt.% to 80 wt.% of [EMIM][TFSI] in p(VDF-HFP) were prepared [127]. The experimental results indicated that the gas permeability strongly increased with an increase in IL concentration and the transport of larger and more condensable species was favored over that of the smaller molecules. For this reason, the selectivities slightly decreased for the gas pairs of CO_2/N_2, CO_2/CH_4 and O_2/N_2 as the IL content increased, whereas CO_2/H_2 selectivity increased greatly with higher IL loadings. Therefore, the ILMs can be used as candidates for applications where CO_2 must be separated from H_2 while the latter remains at high pressure. Subsequently, the transport properties of gases in this kind of QSILMs were studied on the basis of Young's modulus [150]. A transition from diffusion-controlled transport to solubility-controlled transport with the increase of IL content in the ILM was found. Hanioka and coworkers [135] reported that the SILMs prepared with IL based on amino-functionalized imidazolium cation ([C_3NH_2MIM][NTf_2]) exhibited higher CO_2 permeability and lower CH_4 permeability in comparison with the SILM composing of a conventional imidazolium-based IL ([BMIM][NTf_2]). This was mainly attributed to the complex interaction between amines and CO_2. Myers et al. [151] has also found that at 85 °C the CO_2/H_2 selectivity of SILM with [C_3NH_2MIM][NTf_2] was larger than that with [BMIM][NTf_2]. In addition, the SILM with fluoroalkyl-functionalized imidazolium IL was found to exhibit higher selectivity for CO_2/CH_4 and lower selectivity for CO_2/N_2, in comparison with that with alkyl imidazolium analog [137].

In addition, the effect of water content and plasticization behavior of IL immobilized, temperature as well as pressure on the CO_2 separation performance of ILM were also investigated by some investigators (as listed in Table 1) [78,122,157–173]. Zhao and coworkers [157] examined the effect of water content in [BMIM][BF_4] on CO_2/N_2 separation performance of SILMs with PES as support membrane. They found that the CO_2 permeance as well as selectivity of CO_2/N_2 at low water content in [BMIM][BF_4] can be improved due to the increase of CO_2 diffusivity resulted from the decrease of viscosity with increasing the water content. Whereas, the CO_2 permeance decreased at high water content in IL because of a decrease in CO_2 solubility. Neves et al. [117] reported that the water clusters formed inside the support material resulted in the decrease in selectivity of CO_2. The plasticization behaviors of three different polymerized RTILs (poly (RTIL)) at elevated CO_2 pressures were investigated by Simons and coworkers [158]. They found that methane could not plasticize the poly (RTIL) in single gas transport experiments, whereas, the presence of CO_2 can accelerate its transport by more than 250%. The plasticization effect of CO_2 was fully reversible on the time scale of the diffusional processes probably due to the charged nature of the polymer material in comparison with the conventional polymer membranes for high pressure CO_2 separations. Wickramanayake et al. [159] studied the effect of temperature on CO_2 permeance in the hollow-fiber SILMs with [HMIM][NTf_2]. The permeability of CO_2 increased by 4 times as the temperature increased from 37 °C to 200 °C, while the CO_2/H_2 selectivity decreased from 6 to 1. Scovazzo et al. [160] investigated the influence of CO_2 partial pressures on the CO_2-permeability and CO_2/CH_4 selectivity of SILMs with [EMIM] cation-based ILs. It was found that both of the permeability and selectivity remained constant over the CO_2 partial pressure ranging from 0 to 207 kPa and the CH_4 partial

Table 2
Use of ILMs in separation of organic compounds.

Application	IL	Support membrane/LMOG/ polymeric materials	Reference
Separation of transesterification reaction produces	[BMIM][Ac]	Durapore	[193]
	[BMIM][BF_4]	Nylon	[194]
	[BMIM][PF_6]	Fluoropore	[193]
	[BMIM][PF_6]	Isopore	[193]
	[BMIM][PF_6]	Mitex	[193]
	[BMIM][PF_6]	Nylon	[193,194]
	[OMIM][BF_4]	Nylon	[194]
	[OMIM][NTf_2]	Nylon	[194]
	[OMIM][PF_6]	Nylon	[194]
Separation of 1,3-proanediol from water	[N_{3333}][$B(CN)_4$]	NF ceramic membrane	[80]
Separation of 1-butanol from water	[BBIM][BF_4]	PDMS	[186,193]
	[OMA][NTf_2]	PP	[194]
	[Ph_3t][NTf_2]	PP	[194]
	[Ph_3t][DCN]	PP	[194]
Separation of 1-butanol and acetone from water	[EIM][PF_6]	PDMS/TiO_2 ceramic ultrafiltration	[195]
	[EEIM][PF_6]	PDMS/TiO_2 ceramic ultrafiltration	[194]
Separation of 1-butanol and isopropyl alcohol from water	Aliquat 336	PVC/PVDF	[96]
	[BMIM][PF_6]	PTEE	[104]
	Cyphos 101	PVC/PVDF	[96]
	Cyphos 102	PVC/PVDF	[96]
	Cyphos 104	PVC/PVDF	[96]
	Tf_2N-based IL precursor	hydroxyl terminated PDMS	[87]
Separation of lactic acid from the fermentation broth	Aliquat 336	PP hollow fiber	[123]
	Aliquat 336	PVDF	[71,123]
	[BMIM][PF_6]	PP hollow fiber	[123]
	[BMIM][PF_6]	PVDF	[123]
	Cyphos 101	PP hollow fiber	[123]
	Cyphos 101	PVDF	[71,123]
	Cyphos 102	PP hollow fiber	[123]
	Cyphos 102	PVDF	[71,123]
	Cyphos 104	PP hollow fiber	[123]
	Cyphos 104	PTEE	[195]
	Cyphos 104	PVDF	[71,123]
	Cyphos 109	PP hollow fiber	[123]
	Cyphos 109	PVDF	[71,123]
	Cyphos 111	PP hollow fiber	[123]
	Cyphos 111	PVDF	[123]
	[MP Pyperidinium] [$(TMS)_2$N]	PP hollow fiber	[123]
	[MP Pyrrolidinium] [$(TMS)_2$N]	PP hollow fiber	[123]
	[MP Pyperidinium] [$(TMS)_2$N]	PVDF	[123]
	[MP Pyrrolidinium] [$(TMS)_2$N]	PVDF	[123]
Separation of 4-phenoxybutyric, 3-phenoxyproionic acid, 2-phenylpropionic acid, 2-phenoxybutyric acid and 2-amino-2-phenylbutyric acid from aqueous solutions	[BMIM][PF_6]	PP	[72]
	[BMIM][NTf_2]	PP	[72]
	[HMIM][PF_6]	PP	[72]
	[OMIM][PF_6]	PP	[72]
Separation of phenylalanine, phenylglycine and tryptophan from aqueous solutions	[OMIM][PF_6]	PVDF	[82]
Separation of benzene, toluene and p-xylene from aqueous solutions	[BMIM][PF_6]	PVDF	[75]
	[HMIM][PF_6]	PVDF	[75]
	[OMIM][PF_6]	PVDF	[75]
Separation of benzene and cyclohexane	[BMIM][PF_6]	PVDF	[197]
	[HMIM][PF_6]	PVDF	[197]
	[OMIM][PF_6]	PVDF	[197]
Separation of toluene and cyclohexane	[BMIM][PF_6]	PVDF	[198]

(*continued on next page*)

Table 2 (*continued*)

Application	IL	Support membrane/LMOG/ polymeric materials	Reference
Separation of Bisphenol A from aqueous solution	[BMPYR][BTA]	PVDF	[74]
	[BTNH][BTA]	PVDF	[74]
	[C_2DMIM][PF_6]	PVDF	[74]
	[MTONH][Cl]	PVDF	[74]
	[OMIM][PF_6]	PVDF	[74]
	[TBP][PF_6]	PVDF	[74]
	[TBTDP][BTMPP]	PVDF	[74]
	[THTDP][Br]	PVDF	[74]
	[THTDP][Cl]	PVDF	[74]

pressure ranging from 0 to 300 kPa. Experimental results verified that the SILMs are potential to meet the demand of the cross-membrane pressure of less than 2 bar and the CO_2/N_2 selectivities of greater than 50, which may be more efficient than existing systems for CO_2 capture from the flue gases [161,162]. The SILM prepared using PS support membrane and [HMIM][NTf_2] was used to examine the effect of experimental temperature on separation performance of CO_2 from CO_2/He mixture [163]. The CO_2 permeability of the SILM increased from 744 to 1200 barrer as the temperature was elevated from 37 to 125 °C, while the CO_2/He selectivity decreased from 8.7 to 3.1.

4.1.2. Separation of sulfur compounds

Compared to CO_2, sulfur compounds (*e.g.* sulfur dioxide (SO_2) and hydrogen sulfide (H_2S)) are more soluble in some ILs. Separation and concentration of sulfur compounds are required in some processes in order to achieve its recovery and reuse. There are some literature available where ILMs were studied for potential applications in the separation of SO_2/air, SO_2/CH_4, SO_2/N_2 or H_2S/CH_4 [174–177]. Luis et al. [174] investigated the permeabilities of air, CO_2 and 10 vol.% SO_2-air through a SILM with acetate anion-based IL to separate SO_2 or CO_2 from air. It was found that permeability of air was one order of magnitude lower that of CO_2 and also lower than that of 10 vol.% SO_2. The SILM with hydrophilic PVDF membrane as support material and [BIM][AC] as liquid phase showed the highest permeability. The permeability and selectivity of SO_2 through SILMs made of imidazolium-based ILs and the PES membranes were studied at temperatures from 25 to 45 °C [175]. The SILMs can offer very good permeability of SO_2 as well as ideal SO_2/CH_4 and SO_2/N_2 selectivities up to 144 and 223, respectively. And the dissolution-diffusion transport mechanism can be qualitatively applied to analyze the SO_2 permeation. The SILMs, which consist of a solid polymer matrix and [BMIM][BF_4] IL, were prepared for the removal of acidic gases from crude natural gas [176]. The permeability coefficients of CO_2 and H_2S was considerably high at 30–180 and 160–1100 barrer, respectively. The selectivities of CO_2/CH_4 and H_2S/CH_4 were 25–45 and 130–260, respectively, because both of them have higher affinity toward [BMIM][BF_4] IL than CH_4.

4.2. Separation of metals

ILMs have been applied to the separation of metals from aqueous and showed outstanding performances compared to other types of liquid membranes particularly in terms of membrane lifetime [178–185]. The transport of chromium (III) from alkaline aqueous solutions using pseudo-emulsion based hollow fiber strip dispersion (PEHFSD) technology with [N_{8881}]Cl IL as mobile carrier has been studied [178]. Alguacil and coworkers [179] found that saturation of the carriers did not occur, as it was continuously regenerated in the pseudo-emulsion. By the use of the PEHFSD technology, the transport of iron (III) from acidic media was also studied using [PJMTH]$_2$[SO_4] IL as carrier. The results indicated that the mass transfer resistance for the membrane phase was negligible in comparison with the overall mass transfer resistance, and the PEHFSD technology performed well compared to the other hollow fiber technologies. In addition, the extraction of chromium (VI) from aqueous acidic solutions was performed using Cyphos 101 IL as carrier [180] Transport of chromium increases with an increase in carrier concentration, and extraction of chromium up to 95% can be obtained under a given experimental condition. Guo et al. [181] also investigated chromium (IV) transport using QSILM with PVDF as support, [$C_n$mim][PF_6] or [$C_n$mim][BF_4] ($n = 4,8$) as IL plasticizers and Cyphos 104 as carrier. They found that the permeability of QSILM with Cyphos 104 as carrier was 13 times faster than that of Aliquat 336, and the QSILM with [OMIM][BF_4] and Cyphos 104 revealed the better permeability coefficient for chromium (VI) transport. Selective permeation of Dy and Nd ions against Fe ion was conducted by an SILM with PVDF as support material, [OMIM][NTf_2] as the liquid phase and DODGAA as the mobile carrier [182]. Using the SILM, Dy and Nd ions could be selectively recovered to the receiving phase, and Fe ions were transported at the 10% level. From these results, it is clear that the ILMs showed longer reusability and larger flux in comparison with the other SLMs [183–185].

4.3. Separation of organic compounds

Compared to the commonly used methods, *e.g.* liquid-liquid extraction, precipitation, absorption and distillation, ILMs technology for separation of organic compounds does

not require high energy consumption because they can be conducted under moderate operating conditions. As of now, ILMs have been successfully applied for the highly selective separation of organic compounds, *e.g.* transesterification reaction produces [186–191], alcohols [34,36,37,80,87,96,104,104,141,192–194], organic acids [71,72,82,123,195,196], hydrocarbons [74,75,197,198] and amines [76–78]. Table 2 summarized the separated system, the IL, and the corresponding support membrane/LMOG/polymeric materials.

4.3.1. Transesterification reaction products

The application of ILMs for separation of transesterification reaction produces, *i.e.* vinyl butyrate, 1-butanol, butyl butyrate and butyric acid, has been investigated by De los Ríos and coworkers in detail [186–191]. In order to evaluate the effect of the support membrane nature on the selectivity of SILMs, the transport of transesterification reaction products through SILMs with [BMIM][PF_6] immobilized in five different polymeric membranes (Nylon, Isopore, Fluoropore, Mitex and Durapore) was evaluated [186]. The researchers found that the most suitable support membranes for the selective separation of compounds were Nylon and Isopore. To evaluate the effect of nature of ILs on the selectivity of the SILMs, the permeability of the four compounds through SILMs made from six imidazolium-based ILs immobilized in Nylon membranes was measured [187]. It can be concluded that the permselectivity for the four compounds increased with an increase in the hydrophilic character of ILs. The SILM based on [BMIM][BF_4] was the most effective for separating the four compounds. A wide range of ILs with different cation and anion compositions as liquid phase in SILMs were further tested to understand the mechanisms of ILs for selectively separating the compounds [188]. It was found that selectivity values of SILMs are mostly dependent on the anion of IL. For SILMs based on ILs with a given anion, selectivities increased with the decrease in the alkyl chain length on the imidazolium ring. In addition, to elucidate the molecular mechanism involved in the transport phenomena of the target compounds, De los Ríos and coworkers [189–191] established a relationship between the permeability and the partition coefficients based on the Wilke-Chang equation. Based on this equation, the separation capability of a given SILM can be easily predicted by evaluating the partition coefficient and the molar volume of each compound.

4.3.2. Alcohols

The separations of the binary systems (1,3-proanediol-water and 1-butanol-water), the ternary systems (such as 1-butanol-acetone-water and 1-butanol-isopropyl-water) and the quaternary system (such as 1-butanol-acetone-ethanol-water) have been investigated respectively by some researchers using different ILMs. As for the binary 1,3-proanediol-water system, Izák et al. [80,192] studied the selectivity separation of the 1,3-proanediol from aqueous solution using a SILM with a NF ceramic membrane as support and [N_{3333}][$B(CN)_4$] as liquid phase by pervaporation. The separation factor of 1,3-propanediol can be increased from 0.4 to 177 by using the SILM. This may be attributed to the fact that 1,3-propanediol preferentially passed through the SILM with the hydrophobic [N_{3333}][$B(CN)_4$] inside the pores of ceramic membrane.

As for the binary 1-butanol-water system, Kohoutová et al. [86] investigated the separation of 1-butanol from its 5 wt% of aqueous solution using the quasi-solidified [BBIM][BF_4]-PDMS membranes. This kind of membrane showed high stability even if it contained [BBIM][BF_4] and PDMS phases. Moreover, the separation factor of 1-butanol raised up to 37 when 30 wt.% of [BBIM][BF_4] was accommodated in PDMS. Following this work, the effects of quasi-solidified [BBIM][BF_4]-PDMS membranes on the sorption and desorption kinetics of 1-butanol in aqueous solution were investigated [193]. Although anomalous sorption occurred at higher concentrations of 1-butanol, based on the generalized Fick's second law, the calculated values were in good agreement with the measured data of sorption and desorption kinetics. In addition, the pervaporative recovery of 1-butanol from dilute aqueous solutions was carried on using SILMs with a hydrophobic PP flat sheet membrane as support and with ammonium- or phosphonium-based IL as liquid phase [194]. It was found that the selectivities of SILMs increased with the increasing 1-butanol concentration in feed solution probably due to the capacity of 1-butanol to impede water sorption and diffusion. And it can be concluded from the diffusivity and activation energy analyses that water transport was mainly governed by the presence of water microenvironment in SILM. Heitmann et al. found that the permeation properties of 1-butanol could be influenced by immobilization method of ILs [36]. The total permeate flux of QSILM prepared by simple dissolution of [C_{10}MIM][TCB] in PEBA was up to 560 g m^{-2} h^{-1}, and the highest concentration of 1-butanol in the permeate was found to be 55 wt.%. Izák et al. [37] investigated the pervaporation separation properties of 1-butanol-water mixtures through a SILM which was prepared by impregnating a mixture containing [EEMIM][PF_6] and PDMS into an TiO_2 ceramic module. The diffusion coefficients of 1-butanol in the SILM were much higher than in PDMS only.

As for the ternary systems, Izák and coworkers [141] firstly studied the separation of the 1-butanol and acetone from water using the supported [N_{3333}][$B(CN)_4$]-PDMS membrane with ultrafiltration membrane as support material. In comparison with a PDMS membrane, the enrichment factors of 1-butanol and acetone increased from 2.2 to 10.9 and 2.3 to 7.9, respectively, when the supported [N_{3333}][$B(CN)_4$]-PDMS membrane was used. Matsumoto et al. [96] prepared the quasi-solidified IL-PVC membrane to investigate the separation of 1-butanol and isopropyl alcohol. The QSILM with the ratio of Aliquat 336/PVC = 70/30 (wt./wt.) gave the highest permeability and separation factor. A 1-butanol flux of 27 g m^{-2} h^{-1} can be achieved even if its permeate concentration is lower than 10 wt.%. But, the separation factor and selectivity were insensitive to the membrane thickness and feed concentration.

As for the quaternary system, Plaza et al. [104] investigated the separation of 1-butanol-acetone-ethanol (ABE) from aqueous using a QSILM obtained by the gelation of [BMIM][PF_6] into the pores of PTEE hollow fibers. Mai and coworkers [87] fabricated a quasi-solidified IL-PDMS membrane in which a [Tf_2N]$^-$-based IL covalently bound to the PDMS backbone polymer and used it to separate ABE from aqueous solution by pervaporation. The permeate flux of the QSILM was 7.8-times higher than that of conventional SILM in which the [Tf_2N]-based ILs were physically absorbed inside PDMS. In addition, by pervaporation, Izák et al. [34] found that the 1-butanol and acetone solvents could be efficiently removed from the ABE fermentation broth using a SILM with a mixture containing 15 wt.% of [N_{3333}][$B(CN)_4$] and 85 wt.% of PDMS. From these work, it can be concluded that ILMs have the potential for separation of butanol from the ABE solutions by pervaporation, although fluxes of ILMs are generally lower than those of polymeric, ceramic and some other composite membranes.

4.3.3. Organic acid

The technique of ILM has been used as alternative to solvent extraction, adsorption, direct distillation and electrodialysis for the effective and selective isolation and separation of organic acid, such as lactic acid (LA), 4-phenoxybutyric, 3-phenoxyproionic acid, 2-phenylpropionic acid, 2-phenoxybutyric acid, 2-amino-2-phenylbutyric acid, and salicylic acid (SA) [54,71,82,123,195]. Lactic acid (LA), which is the raw material of biodegradable polylactic acid, can be prepared by fermentation. As of now, the production cost of LA is mainly accounted to its separation and purification from the fermentation broth. In order to find an effective and economical downstream process for recovering LA from fermentation broth, Matsumoto and coworkers [123] firstly investigated the effect of IL type on the permeation behavior of LA through SLM based on ILs in detail. They found that the permeation flux of LA through the hollow fiber membrane impregnated with Aliquat 336 was higher than the estimated value with the flat sheet membrane. Subsequently, Aliquat 336, Cyphos 101 and Cyphos 102 were found to be the best membrane solvents in terms of membrane stability and permeation of LA when hydrophilic PVDF was used as support material of SILM [71]. However, in terms of the separation capability of LA, Cyphos 102 was the best. In addition, the mechanism of LA passing through SILMs with Cyphos 104 as liquid phase and PTFE membrane as support material was proposed on the basis of experimental results and equilibrium model [195].

As for other organic acids, in order to verify the concept that an SILM is expected to achieve much faster transport of a substance and reduce the volume of the liquid membrane phase in comparison with the bulk liquid membrane, Miyako et al. [72] have developed a novel SILM system in which lipase-catalyzed reactions facilitated the transport of organic acids, *i.e.* 4-phenoxybutyric, 3-phenoxyproionic acid, 2-phenylpropionic acid, 2-phenoxybutyric acid and 2-amino-2-phenylbutyric acid. There were remarkable differences in the permeate fluxes of various organic acids due to the substrate specificity of the lipases, and the maximum permeate flux of 44×10^{-2} mmol cm^{-2} h was obtained using 4-phenoxybutyric acid as the substrate and [BMIM][PF_6] as the liquid phase. Fortunato and coworkers [82] studied the separation of amino acids, *i.e.* phenylalanine, phenylglycine and tryptophan from aqueous solutions by SILMs based on [OMIM][PF_6]. The extraction of amino acids by the IL were negligible in comparison with those of amino acid esters. This may be attributed to the fact that amino acids show the higher affinity toward water probably due to the hydrogen bond formation between water and the carboxyl group of amino acids [196]. Whereas, amino acid esters are more hydrophobic because the acidic proton of amino acids was substituted by an alkyl group, indicating that SILMs based on [OMIM][PF_6] can be applied to separate amino acid esters from amino acids. The transport of salicylic acid (SA) from aqueous solutions has also been investigated using a SILM with [C_6MIM][PF_6] [54]. The transport efficiency of SA decreased with increasing pH, indicating that the un-dissociated form of SA was mainly extracted.

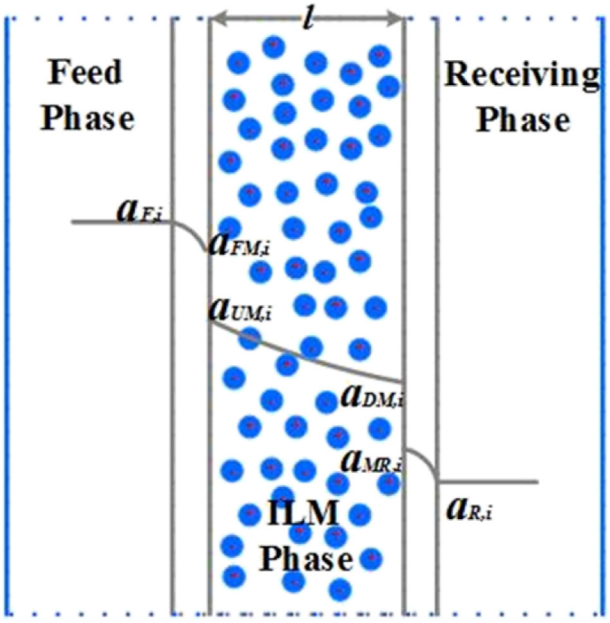

Fig. 4. Scheme of the three-phase system through an ionic liquid membrane.

4.3.4. Aromatic hydrocarbons

The potential of ILMs for the separation of aromatics from aliphatic hydrocarbons was also investigated by several researchers [74,75,197,198]. Selective transport of benzene, toluene and xylene in PVDF membranes containing different ILs has been examined by Matsumoto et al. [75]. Although the permeation rates through SILMs followed the order of the increasing hydrophobicity of the ILs and were lower than those through water-based SLMs because of the high viscosities of the ILs, the selectivity of these aromatic hydrocarbons was greatly improved by the SILMs. SILMs prepared by impregnating imidazolium-based ILs inside the pores of hydrophilic

PVDF have been applied to the vapor permeation process for benzene and cyclohexane separation [197]. The effects of ILs and the benzene fraction in the feed on the permeation flux, separation factor and the stability of the SILMs were investigated. The separation factor depending on the hydrophilicity of IL was mainly affected by the sorption step. A SILM with [BMIM][PF_6] and hydrophobic PVDF membrane was applied for the separation of toluene-cyclohexane mixtures [198]. The separation factor of the SILM reached 15–25 at 40 °C after a 550 h test, and the IL was hardly displaced from the pores of PVDF membrane even under high vacuum conditions. This indicates that the affinity between [BMIM][PF_6] and PVDF membrane resulted in a strong capillary force to hold the IL. SILMs with PVDF membrane and hydrophobic ILs based on phosphonium, imidazolium, ammonium, or pyridinium were used to remove Bisphenol A from aqueous solution [74]. A maximum of 62% permeation of Bisphenol A from feed solution to receiving solution was obtained for the supported [TBP][PF_6]-PVDF membrane. The negligible decrease (<2%) in the SILM weights before and after the transport experiments indicated that the ILs used could not be easily displaced from the pores of PVDF membrane under a cross-membrane pressure gradient.

4.3.5. Amines

Branco et al. [35,76,77] studied the potential of SILMs for selective separation of the organic isomeric amines hexylamine, diisopropylamine and triethylamine in diethylether according to the concentration gradient of the component and its solubility in IL. They found that the appropriate combination of selected RTILs and supporting membranes is crucial for achieving good selectivity in a given separation problem [74]. The SILM with [BMIM][PF_6] as liquid phase and PVDF as support exhibited an extremely highly selective transport of secondary amines over tertiary amines (up to a 55:1 ratio) with very similar boiling points, because of stronger hydrogen bonds of the secondary amine to 2-H of the imidazolium cation. This result indicated that this methodology is feasible for continuous separation of compounds from complex mixtures under simple conditions.

5. Transport mechanisms through ILMs

5.1. Transport mechanism of gases through membranes

The transport mechanism of gases through ILMs is extremely important and has been studied over the last years by a number of groups [85,135,164,172]. It is assumed that the gas transport through a dense liquid membrane occurs according to a solution-diffusion mass transfer mechanism. Generally, a gas molecule is transported across an ILM by the following three steps: (1) absorption in the upstream surface of the ILM, (2) diffusion across the ILM matrix, and (3) desorption in the downstream face of the ILM.

The driving force of a gas molecule across membrane is pressure differential between the feed side and the permeate side. The steady-state flux of gas i through the membrane ($J_{g,i}$) can be determined from the gas permeability ($P_{g,i}$), the membrane thickness (l) and the pressure difference across the membrane ($\Delta p_{g,i}$) according to the following equation:

$$J_{g,i} = \frac{P_{g,i}\Delta p_{g,i}}{l} \tag{1}$$

The ideal gas permeability through an ILM is related to gas solubility and diffusivity data of the ILs:

$$P_{g,i} = S_i D_i \tag{2}$$

Therefore, the ideal selectivity (α_{ij}) is then calculated by taking the ratio of the ideal gas permeabilities for a given gas pair (i and j):

$$\alpha_{ij} = \frac{P_{g,i}}{P_{g,j}} = \frac{S_i D_i}{S_j D_j} \tag{3}$$

The ideal selectivities in Eq. (3) are driven mainly by the differences in the physical solubility of the gases in the IL. The solubility coefficient of gas in IL phase can be calculated using Eq. (4):

$$S_i = \frac{C_{g,i}}{p_{g,i}} \tag{4}$$

At low pressures, $C_{g,i}$ can be determined using pure IL density ρ_{IL} and pure IL molecular weight M_w:

$$C_{g,i} = x_i \frac{\rho_{IL}}{M_w} \tag{5}$$

where x_i is mole fraction of dissolved gas in IL phase. Due to low gas solubility in ILs at low pressures, the mole fraction of dissolved gas in IL is approximately equal to zero. Therefore, Henry's law can be defined as follows:

$$H_i = \lim_{x_i \to 0} \frac{\widehat{f}_i}{x_i} \tag{6}$$

Substitution Eqs. (4)–(6) into Eq. (2) yields Eq. (7):

$$P_{g,i} = \frac{D_i \rho_{IL}}{H_i M_w} \tag{7}$$

5.2. Transport mechanism of organic compounds through ILMs

The mass transport of organic molecules is generally described by the solution-diffusion model, based on which the transport mechanism of organic compounds is considered to be a five-step process: (1) diffusion from the bulk of the feed solution to the upstream side of the membrane, (2) selective sorption at the upstream side of the membrane, (3) diffusion through the membrane, (4) desorption at the downstream side of the membrane, and (5) diffusion away from the membrane on the permeate side [72,195]. The scheme of a three-phase system through ILM is schematically shown in Fig. 4. For pervaporation process, the transport mechanism of organic molecules through an ILM includes only steps 1–4 [70,82,194].

Total flux can be used to evaluate the performance of a membrane. The flux $J_{o,i}$ of component i through the membrane can be described to a first approximation by Fick's Law, which is concentration-based [199]. However, the advantage of using an activity rather than a concentration based expression is that thermodynamic equilibrium is assumed at the interfaces. Under the assumption that the accumulation in both boundary layers and the ILM was neglected because of a very low volume of these layers compared with the volume of the feed and the receiving phases, the mass transport through a dense membrane is expressed by the following equation [200]:

$$J_{o,i} = \frac{P_{o,i}}{l}(a_{F,i} - a_{R,i}) \tag{8}$$

where $a_{F,i} = x_{F,i}\gamma_{F,i}$ and $a_{R,i} = y_{R,i}p_R/p_i^0$ denote the activity of component i in the feed and in the receiving, respectively. The influence of the feed temperature on the permeability parameter can be described by an Arrhenius-type equation:

$$P_{o,i} = P_{o,i}^0 \exp\left[\frac{E_i}{R}\left(\frac{1}{T_0} - \frac{1}{T_F}\right)\right] \tag{9}$$

The two parameters $P_{o,i}^0$ and E_i are evaluated from measurements at several different temperatures. This ideal approach is suitable for binary aqueous systems with low concentrations of organic components but it may be inadequate for higher organic concentrations or multi-component systems.

Another very important factor for the evaluation of the membrane performance is the separation factor, $\beta_{i/j}$, which is defined as:

$$\beta_{i/j} = \frac{w_{R,i}/w_{R,j}}{w_{F,i}/w_{F,j}} \tag{10}$$

The pervaporation separation index (PSI) can be calculated using the following equation:

$$\mathrm{PSI} = J_o(\beta_{i/j} - 1) \tag{11}$$

6. Conclusions

Ionic liquid membranes which combine the advantages of IL with solid support materials have been applied to a wide range of separation processes. The preparation methods of ILM have a significant effect on its stability. Compared to the SILMs, QSILMs are more stable due to the fact that the flowing of ILs can be effectively controlled. However, only a limited number of gels that can quasi-solidify ILs and can be used to prepare for the QSILMs. As of now, ILMs have been successfully used in many fields, *e.g.* separation of mixtures, and can be further expanded into other areas. But, some challenges need to be tackled before the large scale application of ILM technology, such as the compatibility between IL and support membrane, long-term stability of ILM, and scale-up issues.

Author contributions

The paper was written through contributions of all authors. All authors have given approval to the final version of the paper.

Conflict of interest

None.

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Nomenclature

$C_{g,i}$	the concentration of the dissolved gas in IL phase
D_i	the diffusion coefficient
E_i	the active energy
H_i	Henry's law constant
$J_{g,i}$	the steady-state flux of gas i through the membrane
$J_{o,i}$	the flux of organic compound i through the membrane
l	the membrane thickness
$p_{g,i}$	the permeate gas partial pressure
$P_{g,i}$	the permeability of gas i
$P_{o,i}$	the permeability of organic compound i
$\Delta p_{g,i}$	the pressure difference across the membrane
$P_{o,i}^0$	the permeability of component i at the standard state
PSI	pervaporation separation index
R	the gas constant
S_i	the solubility coefficient
T_0	the temperature at the standard state
T_F	the temperature in the feed phase
$w_{F,i}$	the weight fractions of component i in the feed phase
$w_{F,j}$	the weight fractions of component j in the feed phase
$w_{R,i}$	the weight fractions of component i in the receiving phase
$w_{R,j}$	the weight fractions of component j in the receiving phase
$\alpha_{F,i}$	the activity of component i in the feed phase
$\alpha_{R,i}$	the activity of component i in the receiving phase
α_{ij}	the ideal selectivity
$\beta_{i/j}$	the separation factor
$\widehat{f}_i$	the fugacity of the dissolved gas in gas phase
ρ_{IL}	the density of IL

Abbreviations

ILs	ionic liquids
Aliquat 336	Trioctylmethylammonium chloride

Cyphos 101 Trihexyltetradecylphosponium chloride
Cyphos 102 Trihexyl-tetradecylphosponium bromide
Cyphos 104 Tributyltetradecylphosponiumbis(2,4,4-trimethylpentyl)phosphinate
Cyphos 109 Trihexyltetradecylphosphonium bis-trifluoromethylsulfonyl imide
Cyphos 111 Trihexyltetradecylphosphonium tetrafluoroborate
DODGAA N,N-dioctyldiglycol amic acid
EDX energy dispersive X-ray
LMOGs low-molecular-weight organic gelators
PA polyamide
PBI polybenzimidazole
PDMAA poly(dimethylacrylamide)
PDMS polydimethylsiloxane
PEBA poly(ether block amide)
PEG poly(ethylene glycol)
PES polyethersulfone
PI glassy fluorine-containing polyimide
PMDA-ODA PI poly(pyromellitimide-*co*-4,4'-oxydianiline)
PP Polypropylene
PS polysulfone
PTEE polytetrafluoroethylene
PVC polyvinyl chloride membrane
PVDF polyvinylidene fluoride
p(PVDF-HFP) poly (vinylindene fluoride-*co*-hexafluoropropylene)
PVP poly-(vinylpyrrolidone)
QSILM quasi-solidified ionic liquid membrane
RTILs room temperature ionic liquids
SEM scanning electron microscopy
SILM supported ionic liquid membrane
VP 4-vinylpyridine
VPrr 1-vinyl-2-pyrrolidinone

Cations

[APMIM] 1-(3-aminopropyl)-3-methylimidazolium
[Aliquat] tri-C_8-C_{10}-alkylmethylammonium
[BBIM] benzyl-3-butylimidazolium
[BIM] 1-butylimidazolium
[BMIM] 1-butyl-3-mthylimidazolium
[BMP] 1-butyl-3-methylpyrrolidinium
[BMPYR] 1-Butyl-1-methylpyrrolidinium
[BTNH] Butyltrimethylammonium
[C_2DMIM] 1-ethyl-2,3-dimethylimidazolium
[C_3MIM] 1-propyl-3-mthylimidazolium
[C_3NH_2MIM] N-aminopropyl-3-methylimidazolium
[C_8F_{13}MIM] 1-methyl-3-(3,3,4,4,5,5,6,6,7,7,8,8,8-tridecafluorooctyl)imidazolium
[C_{10}MIM] 1-decyl-3-methylimidazolium
[EEIM] 1-ethenyl-3-ethyl-imidazolium
[EEMIM] 1-ethenyl-3-ethyl-imidazolium
[EMIM] 1-ethyl-3-mthylimidazolium
h[MIM]$_2$ (1,6-di(3-methylimidazolium)hexane
[HMIM] 1-hexyl-3-mthylimidazolium
[MIM] 1-methylimidazolium
[MP Pyrrolidinium] N-Methyl-N-propylpyrrolidinium
[MP Pyperidinium] N-Methyl-N-propylpiperidinium
[N_{3333}] tetrapropylammonium
[N_{2224}] triethylbutylammonium
[N_{8881}] trioctylmethylammonium
[OMA] trioctylmethylammonium
[OMIM] 1-octyl-3-methylimidazolium
pr[MIM]$_2$ 1,3-di(3-methyl-imidazolium)propane
[P_{4444}] tetrabutylphosphonium
[Ph_3t] Trihexyl(tetradecyl)phosphonium
[SMMIM] 1-methyl-3-(1-trimethoxysilylmethyl)imidazolium
[TBP] tetrabutylphosphonium
[THTDP] trihexyltetradecylphosponium
[Vbtma] vinylbenzyltrimethylammonium

Anions

[AC] acetate
[$B(CN)_4$] tetracyanoborate
[BETI] bis(perfluoroethyl(sulfonyl))imide
[BF_4] tetrafluoroborate
[Br] bromide
BTA Butyltrimethylammonium
BTMPP bis(2,4,4-trimethylpentyl)phosphinate
[$C(CN)_3$] tricyanomethane
[CF_3SO_3] trifluoromethanesulfone
[Cl] chloride
[DCA] dicyanamide
[dimalonate] dimalonate
[diglutarate] diglutarate
[dimaleate] dimaleate
[ESO_4] ethylsulfate
[Gly] glycine
[Lac] lactate
[malonate] malonate
[maleate] maleate
[MSO_4] methylsulfate
[NTf_2] bis(trifluoromethylsulfonyl)imide
[OTf] trifluoromethanesulfonate
[PF_6] hexafluorophosphate
[Pro] proline
[SCN] thiocyanate
[TCB] tetracyanoborate
[TCM] tricyanomethanide
[TfA] trifluoroacetate
[TFSI] bis(trifluoromethylsulfonyl)
[$(TMS)_2N$] Bis(trifluoromethanesulfonyl)imide

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